ACCUMULATION OF FALLOUT RADIOISOTOPES BY BIVALVE MOLLUSCS

FROM THE LOWER TRENT AND NEUSE RIVERS

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Abstract. Bivalve molluscs have proven useful indicators of the distribution of fallout radioisotopes in an aquatic environment. Three species of filter-feeding molluscs, *Elliptio complanatus, Rangia cuneata,* and *Polymesoda caroliniana,* were collected from stations in the Trent and Neuse Rivers in Eastern North Carolina at 4- to 6-week intervals for more than a year. The soft tissues of each species were ashed and analyzed for gamma radioactivity with a multichannel spectrometer and 4×4-inch NaI crystal. The concentrations of gamma-emitting fallout radioisotopes were monitored in *Rangia* over a 30-mile stretch of river and a salinity range of <0.1 to $>15\%_{oo}$. Ruthenium-106, ^{144}Ce, ^{137}Cs, ^{54}Mn, and ^{65}Zn were present in all samples. After the Chinese nuclear tests in May and December 1966, the amounts of ^{141}Ce, ^{103}Ru, ^{95}Zr-^{95}Nb and ^{140}Ba-^{140}La rose suddenly in the samples. Ruthenium-106 and ^{103}Ru were more concentrated in *Rangia* from downstream stations (salinity range $6-15\%_{oo}$), whereas ^{137}Cs was more abundant in the same species from fresher water (salinity range $0-8\%_{oo}$). Zirconium-95—niobium-95 from the Chinese test in May remained in *Rangia* from freshwater stations after it was no longer detectable in the same species collected downstream.

Introduction

Considerable quantities of radioactive materials have been deposited on the earth's surface as a result of fallout from nuclear weapons tests. Radioisotopes thus introduced into the environment have proven useful as tracers for the study of the biogeochemical cycling of certain elements. Bowen and Sugihara (1965) described their studies of fallout distribution in the Atlantic Ocean as a "geochemical tracer experiment." Since the moratorium on atmospheric testing of nuclear weapons, worldwide fallout has decreased greatly, but long-lived radioisotopes from the huge reservoir of the stratosphere will continue to fall upon the earth's surface for several years. Concentrations of ^{137}Cs and ^{90}Sr, with half-lives of 30 and 28 years, are in fact still increasing on the earth's surface (United Nations 1962). This deposition will be supplemented by fallout of shorter-lived radioisotopes from recent atmospheric tests by nations not participating in the test ban treaty. The periodic introduction of fresh fallout radioactivity into aquatic environments provides unique opportunities for studying the relationships between aquatic organisms and the chemical elements present in the water.

The estuarine environment receives fallout radioactivity from three sources: direct fallout upon the surface of the estuary; freshwater runoff containing radioisotopes leached from the land masses; and tidal exchange of oceanic water within the estuary. The relative magnitudes of these three sources have not been determined for any given estuary. The rate of atmospheric fallout is at least partially dependent upon rainfall (Hardy and Alexander 1962) so that during a rainy season, freshwater runoff probably deposits more radioactivity in the estuary. The contribution of ^{137}Cs from the land mass to freshwater drainage is probably negligible, however, because of the high sorptive capacity of the terrestrial earth for cesium (Davis 1963). Libby (1956)

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found in various rivers less than 5% of the ^{90}Sr concentration estimated for the rain waters supplying the streams, indicating strong terrestrial absorption of ^{90}Sr. Other fission products and neutron-induced radioisotopes might be expected to be equally or more strongly absorbed. Although fallout is greater over the oceans than over the land mass (Polikarpov 1966), the net effect of oceanic mixing in the estuary is probably to dilute the radioactivity present. The shallowness of estuaries, coupled with turbulence from tidal currents, serves to promote absorption of radioisotopes to estuarine sediments. In addition, certain chemical species dissolved or suspended in freshwater runoff tend to flocculate and settle when mixed with seawater in an estuary. In these ways, estuaries act as traps or repositories for fallout isotopes. Estuaries also serve as the habitats for many organisms which are biological concentrators of trace elements and which are therefore subject to radioactive contamination from fallout.

Filter-feeding molluscs are very effective elemental concentrators (Vinogradov 1953, Brooks and Rumsby 1965) useful as indicators of radioactive contamination. Clams (*Mercenaria mercenaria*), oysters (*Crassostrea virginica*), scallops (*Aequipecten irradians*), and mussels (*Modiolus demissus*) from the estuaries near Beaufort, N.C., all accumulated detectable levels of several fallout radioisotopes during 1963–66 (Schelske *et al.* 1964, Schelske 1966, Schelske *et al.* 1967). Bay scallops were especially effective in concentrating ^{54}Mn. Cobalt-60 was concentrated in the soft tissues of the killer clam (*Tridacna gigas*) after nuclear blasts in the Pacific Ocean (Weiss and Shipman 1957). Freshwater mussels (Unionidae) are well-known concentrators of manganese (Vinogradov 1953), and *Unio mancus elongatulus* accumulated ^{54}Mn from fallout in Italy's Lake Maggiore (Ravera and Gaglione 1962, Gaglione and Ravera 1964). In the Columbia River, another unionid mussel, *Anodonta wahlamatensis*, concentrated the induced nuclides ^{54}Mn and ^{65}Zn (Johnson *et al* 1966). The accumulation of ^{54}Mn and ^{65}Zn by freshwater clams has also been discussed by Harrison (1968).

China conducted her third, fourth, and fifth nuclear tests during 1966 — on May 9, October 27, and December 28. According to the U.S. Atomic Energy Commission (1966–1967), each of these nuclear devices employed enriched uranium as the fissionable material. Thermonuclear material was present in the third and fifth tests, but not in the fourth. The yields of the three tests were: May 9 — "in the lower end of the intermediate range (200–1000 kilotons)"; October 27 — "in the low to low-intermediate range"; and, December 28 — "a few hundred kilotons." Only the last test (December 28) was cited as an atmospheric explosion. Freely *et al.* (1966, 1967) detected radioactive debris from the third test (May 9) in the stratosphere, but evaluation as to whether the test occurred in the atmosphere or on the ground proved inconclusive. In the present study we have examined fallout radioactivity from these and previous tests in bivalve molluscs, *Elliptio complanatus* Dillwyn, *Rangia cuneata* Gray, and *Polymesoda caroliniana* Bosc, from the fresh and brackish water of the lower Trent and Neuse Rivers in eastern North Carolina.

The Study Area

The Neuse River drains some 6000 square miles of the central Piedmont region and Coastal Plain of eastern North Carolina (Fig. 1). The only source of radioactive pollutants is from worldwide fallout, and heavy metal pollution by industry is minimal or non-existent. The Trent River enters the Neuse at New Bern, and drains about 500 square miles which consist largely of unpopulated swamps and pocosins with a mean elevation less than 50 ft above sea level. The gradient on the rivers within the study area is essentially zero and the tides influence the water level at the Pollocksville station on the Trent River, although the water there remains fresh. The normal tidal fluctuation is 6 inches or less at Pollocksville and about $1–1\frac{1}{2}$ ft at Cedar Point. Although the current in the Trent is very slow and the river bottom is generally covered with debris, i.e., leaves and branches, particle size determinations on 6-cm core samples from each station except Cedar Point showed 96–99% sand. Coarse silt (0.005–0.05 mm diameter) made up the remainder, except for the Pinecliff core, which consisted of 99% sand and 1% clay (<0.002 mm diameter).
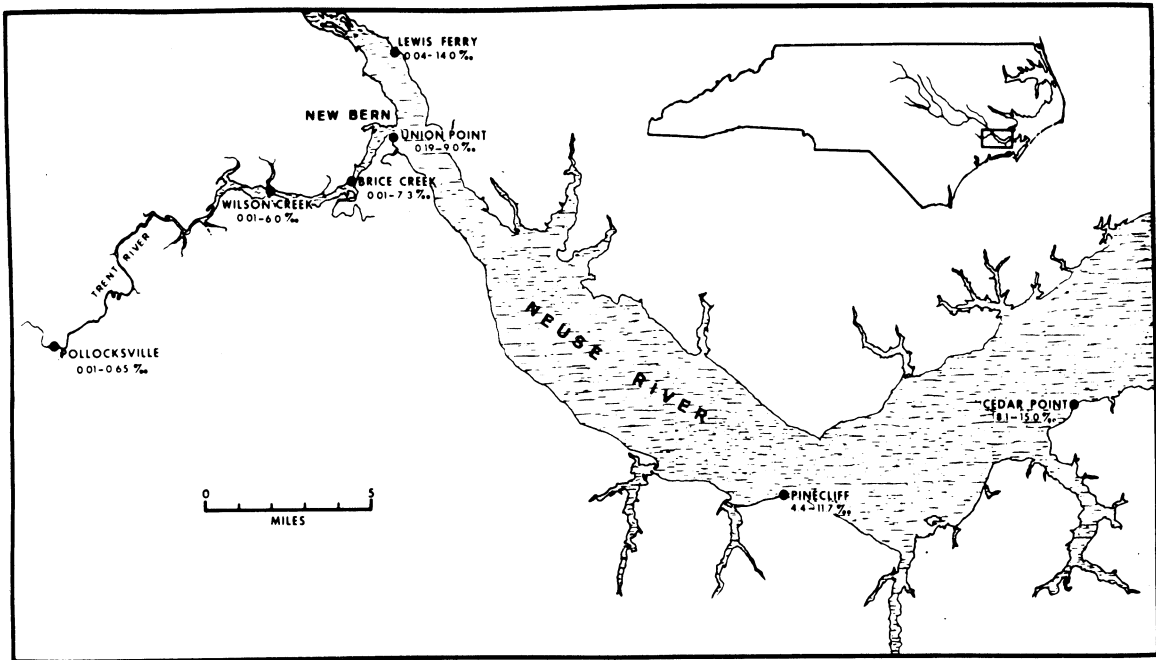
Fig. 1. Sampling stations on the Trent and Neuse Rivers in eastern North Carolina. Salinities shown are the extremes measured at each station during the sampling period. *Elliptio complanatus* was collected only at the Pollocksville station, *Polymesoda caroliniana* mainly from Brice Creek, and *Rangia cuneata* from all stations except Pollocksville.

The total sampling area involved a stretch of river some 42 miles long, and *Rangia cuneata* was collected over a 30-mile stretch with a total salinity range of <0.1 to $>15\%_{oo}$. Rainfall during 1966 was 50–55 inches within the sampling area, and was generally somewhat greater during the late spring and summer months. In the wintertime, therefore, the salinity at Union Point, Lewis Ferry, Brice Creek, and Wilson Creek rose with the decreased runoff, reaching a maximum around December and then falling off to essentially fresh water during April–August. The extremes of salinity measured at each station are shown on Fig. 1. Water temperatures were similar at all stations, and the Union Point station measurements ranged from 5.8 C (2/8/66) to 31.9 C (7/5/66).

Several pelecypods occur within the sampling area. *Elliptio complanatus* was collected at Pollocksville (Fig. 1) and is relatively abundant both upstream and about 5 miles downstream in sand. *Anodonta cataracta*, *Anodonta imbecillis*, and *Lampsilis cariosa* are occasionally encountered with *Elliptio* and are found also farther downstream (to Wilson Creek). *Rangia cuneata* is abundant from a few miles upstream from Wilson Creek in the Trent to the vicinity of Cedar Point, where it is only sparsely distributed. This species is harvested commercially on a limited scale in some estuarine areas of North Carolina. *Polymesoda caroliana* occurs uncommonly from Wilson Creek to Union Point and Lewis Ferry, and was most abundant at the Brice Creek station. *Congeria leucophaeta* occurs over approximately the same range as *Polymesoda*. At the downstream stations, Pinecliff and Cedar Point, *Macoma phenax* is also abundant. *Elliptio*, *Rangia*, and *Polymesoda* were collected at irregular intervals (4 to 6 weeks) from September 1965 through February 1967. Collection at the Cedar Point station was discontinued in the fall of 1966 because of the scarcity of *Rangia*.

Methods and Materials

Collection, Preparation, and Analysis of Samples. Clams were collected by raking. The shells were scrubbed and rinsed at the laboratory, and the animals were steamed in a stainless

steel bucket on a hot plate until the valves gaped. The soft parts and the liquor were separated from the shells, dried at 100 C for 24 hr and weighed. The dried tissues were then ashed at 450 C for 24 hr. The ash was weighed, then loosely packed in a plastic container to either a 50- or a 100-ml volume. Small samples (less than 50 ml ash) were diluted to volume with sawdust, which contained no appreciable radioactivity. Samples were placed directly on a 4 × 4 inch NaI(Tl) scintillation crystal in a low background shield (Schelske 1966) and counted for at least 200 min with a 512 channel analyzer. Only 127 channel capability (20 kev per channel) was used in this study to facilitate treatment of the data.

Analysis of Data. Gamma spectra were stripped manually on the basis of seven standards: [40]K, [65]Zn, [54]Mn, [95]Zr-[95]Nb, ^{137}Cs, [106]Ru, and [144]Ce. Isotopes were determined from the sum of counts in five or six channels for each photopeak. Some samples when first counted obviously contained in addition [140]Ba-[140]La, but this short-lived isotope was permitted to decay before the samples were recounted and the spectra were stripped for the above isotopes. Samples were also recounted at various intervals to determine the proportions of [141]Ce and [144]Ce and of [103]Ru and [106]Ru. The presence of any radionuclides other than the above seven introduces error into the analysis of the entire spectrum. Very minor photopeaks at 1.17 and 1.33 Mev were sometimes found in gamma spectra of *Rangia*, indicating that ^{60}Co from fallout was present in the organism. Unidentified radionuclides in *Rangia* also exhibited small photopeaks in the 0.20–0.35 Mev region of the spectrum. Although [141]Ce and [103]Ru cannot be distinguished from [144]Ce and [106]Ru, respectively, by a single analysis, half-life studies on the cerium and ruthenium photopeaks showed both [141]Ce and [103]Ru to be present in *Rangia* samples collected shortly after the Chinese tests on May 9 and December 28. Stripping the spectra on the basis of [144]Ce and [106]Ru in the presence of [141]Ce and [103]Ru may result in a low estimate of [137]Cs. On analysis of environmental samples, the composite of the stripped standard spectra corresponded well with the experimental count at the photopeak energies, but the composite spectrum is somewhat lower than the experimental spectrum between the photopeaks, indicating the presence of other gamma-emitters in the samples. For *Rangia* samples, the considered radionuclides accounted for 90–95% of the total gamma activity in the samples. Activities reported are those for date of collection and have not been corrected for decay after tests.

Results and Discussion

Apprasial of Gamma Spectra from Elliptio, Rangia, *and* Polymesoda. Certain qualitative differences in the gamma spectra of the three bivalves are immediately apparent (Fig. 2). The *Elliptio* spectrum contained several photopeaks (especially prominent peaks at 2.20, 1.76, and 0.61 Mev) which characterized the spectrum of naturally occurring [226]Ra and daughter products. The presence of [226]Ra in *Elliptio* complicated data analysis and *Elliptio* spectra were not stripped. Of the fallout isotopes, only [106]Ru and [54]Mn are positively identifiable in *Elliptio*, and in fact [54]Mn is the predominant radioisotope concentrated by this organism. Cesium-137 and [65]Zn are probably also present, although the overlapping spectrum of [226]Ra prevents positive identification of either of these isotopes. Except for the appearance and subsequent disappearance of a photopeak at 1.60 Mev ([140]Ba-[140]La), the spectra for *Elliptio* remained relatively unchanged through the Chinese test of May 9 (Fig. 3). Unfortunately, the Pollocksville station was not sampled very soon after the test.

The spectrum of *Polymesoda* (Fig. 2) consists of low counts, mainly because of the small sample size. Traces of [226]Ra may also be present in *Polymesoda*. Like *Elliptio*, *Polymesoda* contains [106]Ru, [54]Mn, and probably ^{137}Cs and [65]Zn. Because of the small samples available and the corresponding low counts, spectra of *Polymesoda* were not analyzed further.

The spectrum of *Rangia* from the Brice Creek station (Fig. 2) shows little or no [226]Ra, but has significant photopeaks from [141-144]Ce, [103-106]Ru, ^{137}Cs, [95]Zr-[95]Nb, and [65]Zn. A small photopeak corresponding to [54]Mn was apparent in *Rangia* spectra before the May 9 Chinese test
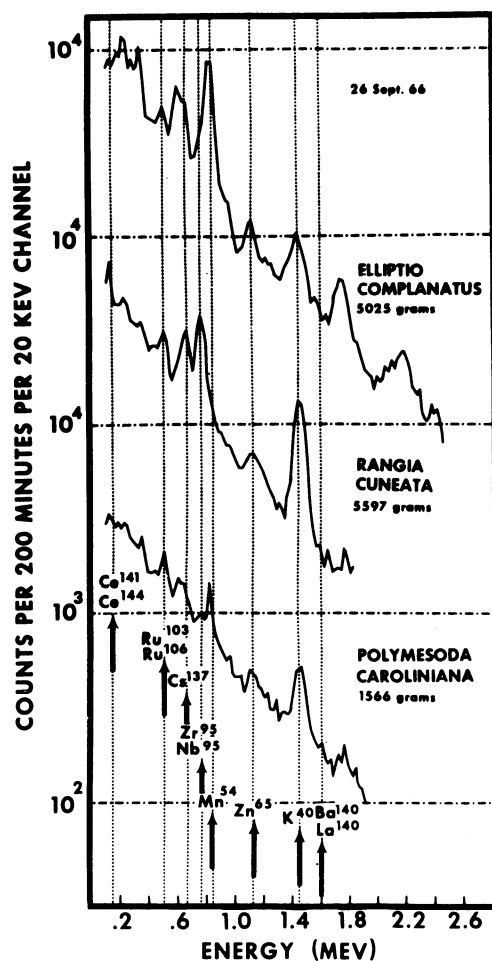
Fig. 2. Gamma spectra of the soft parts of *Elliptio*, *Rangia*, (Brice Creek), and *Polymesoda* collected September 26, 1966. Wet weight of tissue is shown. Three logarithmic scales overlap on the ordinate, and only the highest orders of magnitude are labeled for the top two scales.

(Fig. 3), but this peak was masked by that of ^{95}Zr-^{95}Nb after the test. Zirconium-95—niobium-95 and ^{140}Ba-^{140}La both appeared in spectra of *Rangia* collected May 18, 1966, just 10 days after China's test. Cesium-137 appears (Fig. 3) more concentrated in upstream (Wilson Creek) samples of *Rangia* than downstream (Cedar Point), and conversely the $^{103-106}Ru$ photopeak is relatively larger in the downstream samples. Zirconium-95—niobium-95 had disappeared from clams at Cedar Point (and at Pinecliff) by August 17 (Fig. 3), even though clams from the upstream stations still retained significant quantities of the nuclides. The above relationships are seen more clearly for *Rangia* in the following discussion of the stripped spectra.

Long-Lived Fallout Radionuclides in Rangia cuneata. Five long-lived gamma-emitters from fallout were detected in the soft tissues of *Rangia cuneata* (Table 1). The stations are arranged in downstream order from top to bottom. Cerium-144 is the dominant long-lived fallout radionuclide accounting for at least 40% of the total gamma activity. No significant trends are apparent in the distribution of ^{65}Zn, ^{54}Mn, or ^{144}Ce along the river. Cesium-137 content, however, decreased with the downstream order of the stations. Since the biological uptake of cesium is directly related to that of potassium (Bryan 1963) and since *Rangia* must maintain similar intracellular potassium concentrations in the fresh water at Wilson Creek and in the dilute seawater at

Table 1. Long-lived gamma fallout radioisotopes in Rangia cuneata

Station	Number of samples	Picocuries per 100 grams wet weight of soft parts[1]					
		Zn^{65}	Mn^{54}	Cs^{137}	Ru^{106}	Ce^{144}	Total
Wilson Creek	11	1.27 ± 0.77	5.72 ± 1.77	4.38 ± 1.92	5.12 ± 1.41	11.4 ± 3.12	27.9 ± 4.37
Brice Creek	13	1.40 ± 0.95	7.89 ± 3.07	3.65 ± 1.47	5.36 ± 1.38	15.5 ± 4.41	33.8 ± 5.82
Lewis Ferry	12	1.04 ± 0.91	4.69 ± 2.67	2.74 ± 1.38	4.25 ± 2.11	20.3 ± 8.14	33.0 ± 8.98
Union Point	12	1.18 ± 1.46	5.82 ± 2.02	2.37 ± 1.07	3.97 ± 1.41	11.2 ± 4.70	24.5 ± 5.60
Pinecliff	12	0.72 ± 0.51	7.97 ± 3.86	1.71 ± 0.31	8.00 ± 3.08	11.4 ± 3.22	29.8 ± 5.93
Cedar Point	9	0.80 ± 0.68	7.93 ± 4.91	1.70 ± 0.53	8.16 ± 1.79	16.5 ± 3.86	35.1 ± 6.55
All Stations	69	1.08 ± 0.94	6.65 ± 3.15	2.79 ± 1.55	5.75 ± 2.51	14.2 ± 5.78	30.5 ± 7.27

[1] Means and standard deviations for all samples collected between September 1965 and February 1967.
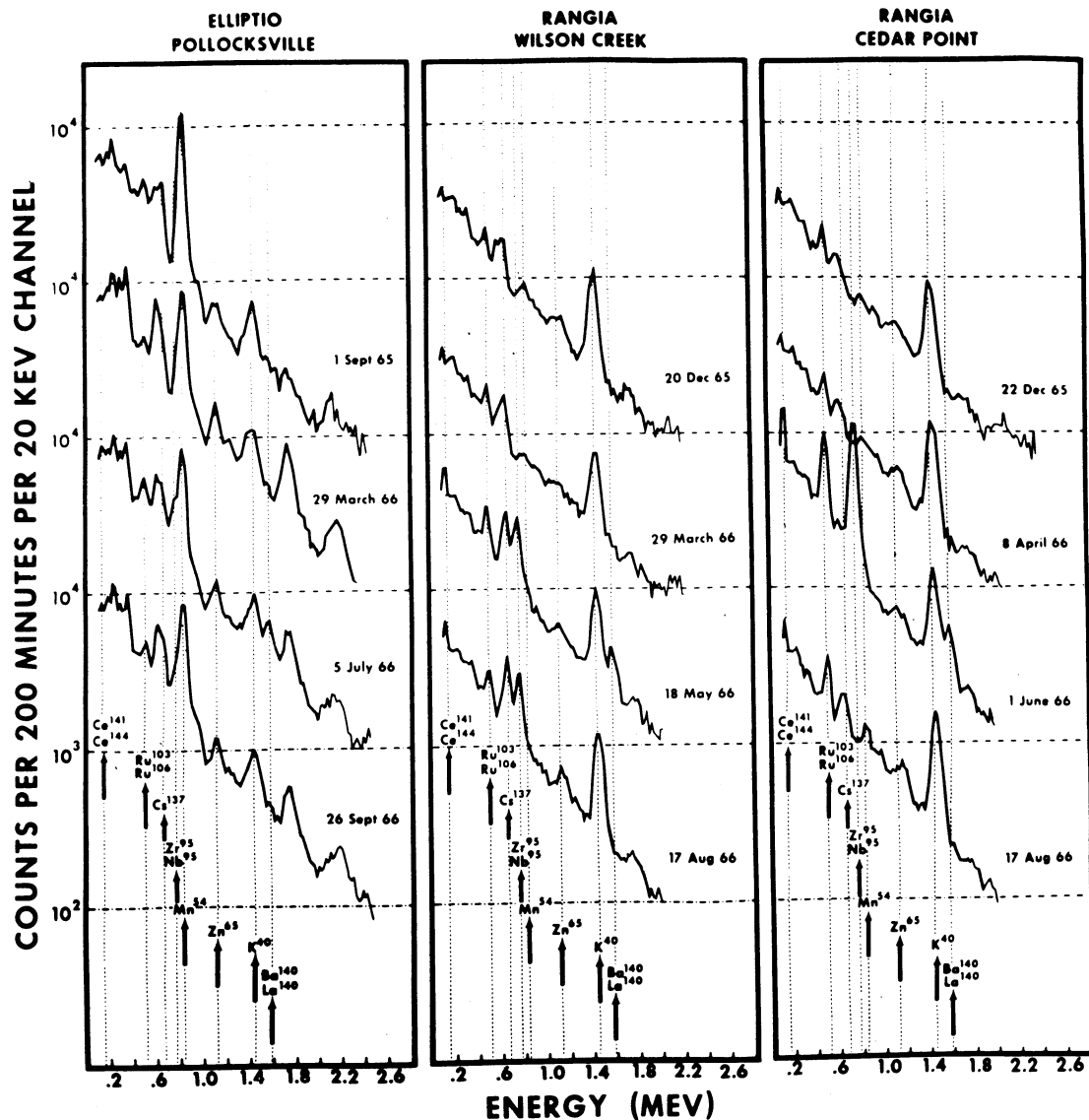
Fig. 3. Gamma Spectra of the soft parts of *Elliptio* and *Rangia* collected during 1965—1966 and showing differences between upstream (Wilson Creek) and downstream (Cedar Point) samples of *Rangia*. Four logarithmic scales overlap on the ordinate, and only the highest orders of magnitude are labeled for the top three scales.

Cedar Point, ^{137}Cs uptake would be suppressed by chemical competition from the higher dissolved potassium concentrations at the downstream stations. Cesium-137—potassium-40 ratios varied inversely with the salinity at each station (Wolfe 1967). Average ^{106}Ru content at the two downstream stations, Cedar Point and Pinecliff, was almost twice that the upstream stations. If ^{106}Ru exhibits a highly solubility and mobility in fresh water (Auerbach and Olson 1963) it was probably less available for uptake by *Rangia* at the upstream stations. In the more brackish waters downstream, ^{106}Ru may be adsorbed onto precipitating ferric hydroxide particles (Jones 1960) which could be directly filtered by *Rangia*.

The range of total long-lived fallout activity was from 11.6—43.9 pCi/100 g wet weight of tissue at the 4 stations shown in Fig. 4. The fluctuations at the upstream stations, Wilson
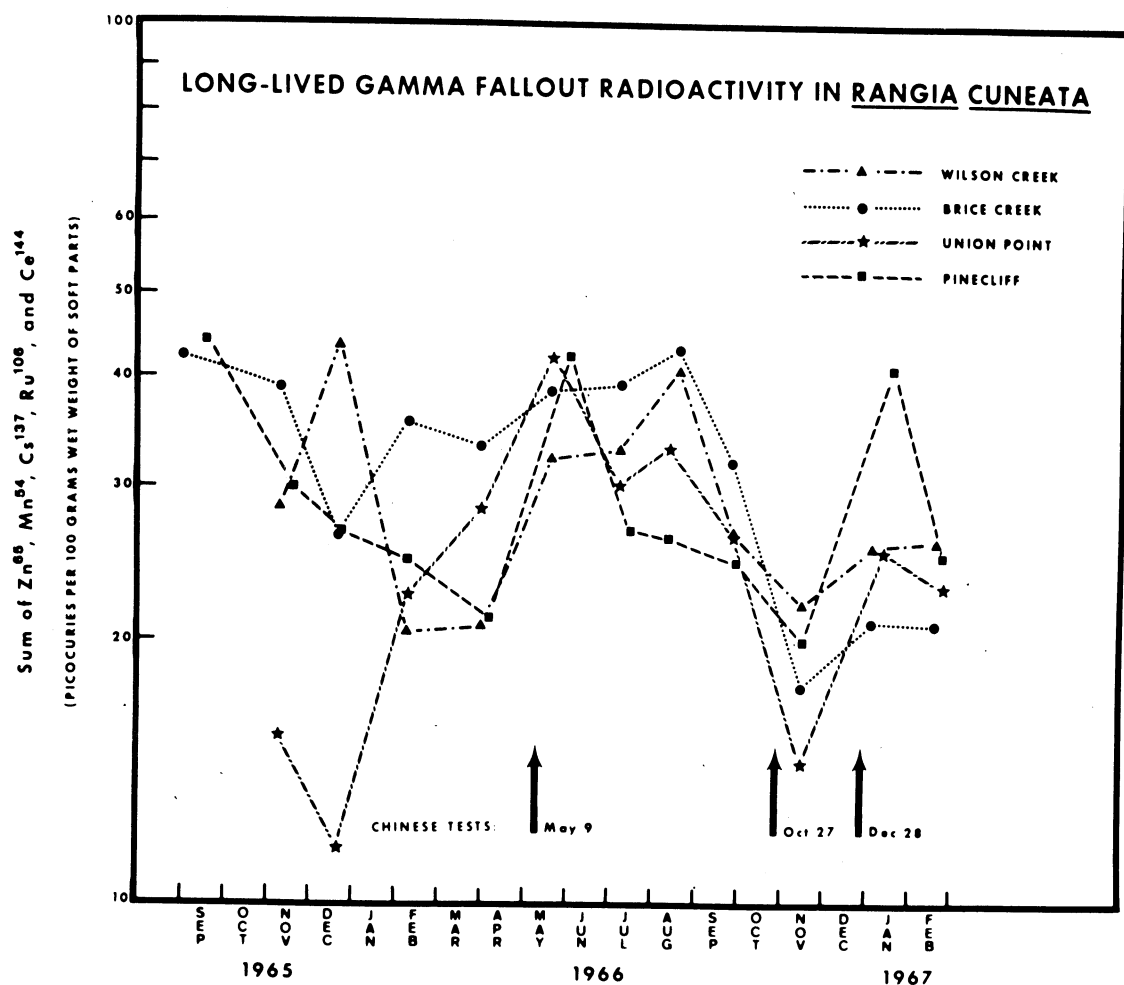
Fig. 4. Content in *Rangia cuneata* of gamma radioactivity from long-lived fallout radionuclides. Stations are labeled from top to bottom in downstream order.

Creek, Brice Creek, and Union Point, appear unrelated to the Chinese tests, and may suggest a seasonal variation with a maximum in midsummer and a minimum during winter. At Pinecliff, on the other hand, the content of long-lived radioactivity abruptly increased after the tests on May 9 and December 28, 1966. These peaks were generated primarily by increases in the relative amounts of ^{106}Ru and ^{144}Ce in the organisms.

Fresh Fallout Radioactivity in Rangia cuneata *from the 1966 Chinese Tests.* The concentrations of gamma-emitting fallout radionuclides measured in *Rangia* were greatest at each station on the first sampling date after the Chinese test on May 9 (Table 2). Unfortunately, the concentrations cannot be compared directly because the two downstream stations were sampled 2 weeks after the four upstream stations. Certain features of the radionuclide distribution are nevertheless outstanding. As discussed previously, ^{137}Cs content decreases with downstream order of the stations, and conversely, ^{106}Ru content is higher at the downstream stations. The distribution of ruthenium in the river is further exemplified by the presence of ^{103}Ru, which was about 10 times as concentrated in downstream *Rangia* on June 1 as in upstream *Rangia* on May 18. The lower content of ^{140}Ba-^{140}La in the downstream samples is probably the result of physical decay of the radioisotope during the 14-day interim between collection of samples.

Table 2. Fallout in Rangia cuneata after Chinese nuclear test - May 9, 1966

Isotope	Wilson Creek May 18	Brice Creek May 18	Lewis Ferry May 18	Union Point May 18	Pinecliff June 1	Cedar Point June 1	Means May 18	Means June 1	Means All
			Picocuries per 100 grams wet weight of soft parts[1]						
Zn^{65}	1.91 ± 0.46	1.77 ± 0.53	1.89 ± 0.25	4.96 ± 0.47	0.49 ± 0.41	1.94 ± 0.34	2.63 ± 1.55	1.22 ± 1.02	2.16 ± 1.48
Mn^{54}	5.27 ± 0.86	6.13 ± 0.94	7.77 ± 0.58	8.97 ± 0.58	11.5 ± 0.80	4.18 ± 0.28	7.04 ± 1.64	7.84 ± 5.18	7.30 ± 2.68
$Zr^{95}-Nb^{95}$	104 ± 8.40	251 ± 10.9	399 ± 6.54	572 ± 7.38	127 ± 7.54	463 ± 8.06	332 ± 200	295 ± 238	319 ± 189
Cs^{137}	8.33 ± 0.15	4.49 ± 0.16	4.55 ± 0.09	4.15 ± 0.09	2.13 ± 0.12	2.62 ± 0.12	5.38 ± 1.97	2.38 ± 0.35	4.38 ± 2.18
Ru^{103}	4.74 ± 1.87	13.2 ± 1.51	6.04 ± 0.99	12.6 ± 0.68	117 ± 1.22	87.1 ± 1.81	9.15 ± 4.37	102 ± 21.2	40.1 ± 66.6
Ru^{106}	5.67 ± 0.93	6.98 ± 1.08	4.66 ± 0.39	2.44 ± 0.54	13.1 ± 0.89	10.6 ± 0.77	4.94 ± 1.90	11.9 ± 1.77	7.24 ± 3.94
Ce^{141}	2.91 ± 2.54	19.4 ± 1.89	35.4 ± 2.11	29.1 ± 0.85	17.3 ± 1.01	60.2 ± 2.22	21.7 ± 13.5	38.8 ± 31.2	27.4 ± 19.5
Ce^{144}	11.3 ± 1.12	19.1 ± 1.31	26.2 ± 0.71	21.4 ± 0.70	15.0 ± 1.04	22.5 ± 0.93	19.5 ± 7.28	18.8 ± 5.30	19.3 ± 6.90
$Ba^{140}-La^{140}$	72.8 ± 3.92	95.6 ± 9.76	172 ± 6.43	124 ± 3.30	48.9 ± 2.33	55.7 ± 2.48	116 ± 42.7	52.3 ± 4.81	94.8 ± 45.8
Total	217 ± 9.95	418 ± 15.0	658 ± 9.52	780 ± 8.24	352 ± 8.22	708 ± 9.00	518 ± 247	530 ± 252	522 ± 224

[1] Deviations shown for individual stations (Columns 2-7) are one sigma counting errors based on the counting rates at the appropriate photopeak energies of the sample and background. Columns 8-10 show the means and standard deviations for the appropriate stations in Columns 2-7.
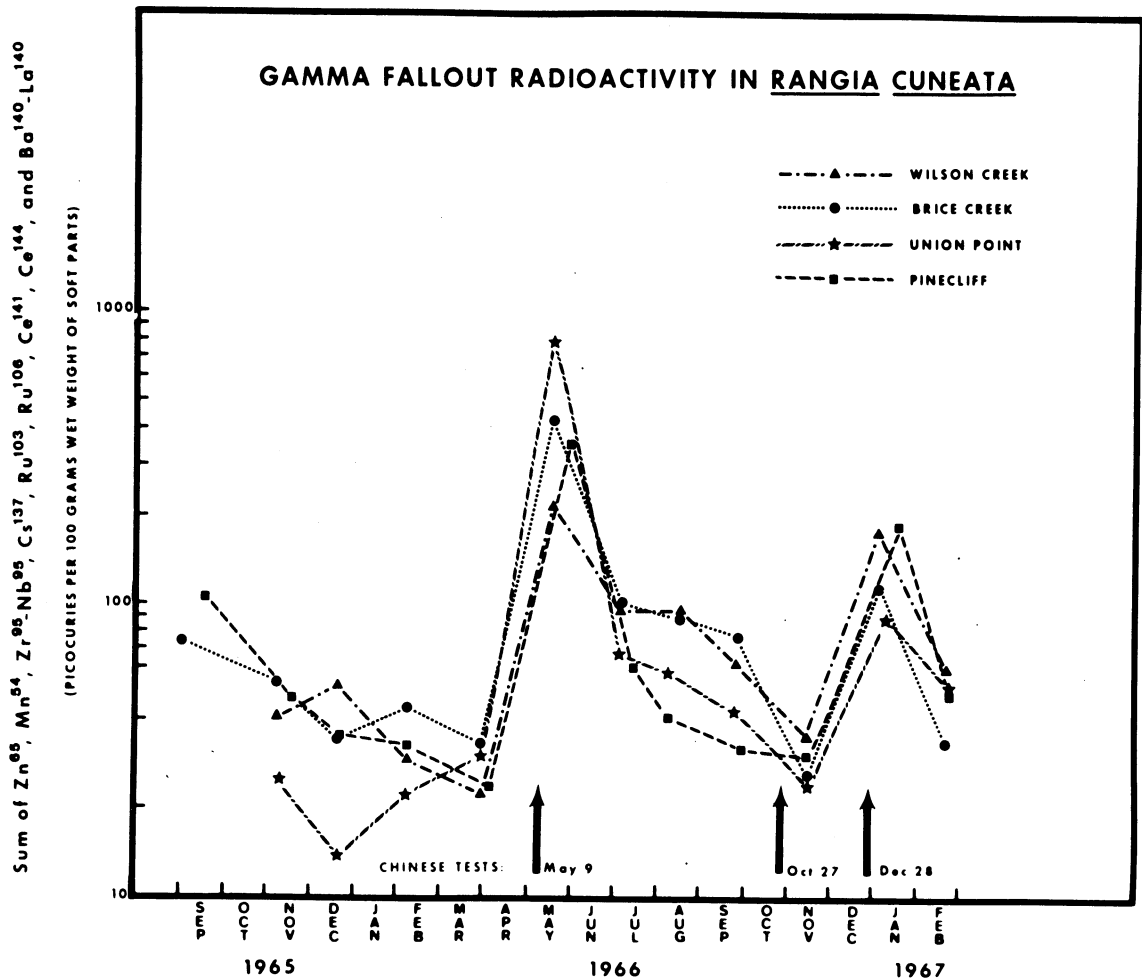
Fig. 5. Content in *Rangia cuneata* of total gamma radioactivity from fallout radionuclides, showing appearance and retention of fallout from Chinese tests. Stations are labeled from top to bottom in a downstream order.

The more rapid disappearance of activity from clams at downstream stations is apparent in Fig. 5. *Rangia* from Wilson Creek, Brice Creek, and Union Point retained higher levels of radioactivity, mainly ^{95}Zr-^{95}Nb, after the initial peak from the May 9 explosion than did clams from Pinecliff. This situation was not apparent from September 1965 to March 1966, however, when Pinecliff clams contained as much or more activity as those from other stations. During this earlier period of sampling, the organisms still retained some residual ^{95}Zr-^{95}Nb from earlier tests (probably from the second Chinese test on May 14, 1965).

The gross gamma activity from fallout in *Rangia* was 7–27 times greater on May 18 than before the Chinese test of May 9 (Fig. 5). A similar increase occurred after the December 28 test. The peaks of radioactive content shortly after the May 9 and December 28 tests consisted mainly (about 80%) of ^{95}Zr-^{95}Nb and ^{140}Ba-^{140}La (Table 2). Thus radioactive debris from the third and fifth Chinese nuclear tests was present in *Rangia* just 10 days and 8 days after the blasts. Samples collected in mid-November 1966 did not reflect a sudden environmental influx of fallout from the October 27 test. This difference could readily be attributed to tests in the atmosphere on May 9 and December 28 versus a surface explosion on October 27. Fallout from the Chinese nuclear tests conducted in 1964 and 1965 reached surface air at New York City initially 8 to 10

days after the tests, and in peak concentrations 12 to 13 days after the tests (Krey and Rosa 1965). If similar circumstances of global transport and fallout prevailed for the third and fifth tests, then movement of fallout radioactivity through the aquatic environment to *Rangia* must be very rapid, and the concentrations of radioactivity detected in *Rangia* May 18, 1966, and January 4, 1967, were not yet at their respective maxima after the two tests. Interception of stratospheric debris from the third test by project Stardust (Feely *et al.* 1966, 1967) was undoubtedly several days after the initial passage of the radioactivity over the U.S. The frequency of sampling for *Rangia* was too low, however, to demonstrate either the actual initial appearance of fresh fallout or the maximum concentrations attained in the clams.

Biogeochemical Considerations of Accumulation. Several mechanisms of uptake by the clams are possible, including direct filtering of insoluble fallout particles, filtering of sediment-sorbed radioactivity, filtering of food organisms after a preliminary planktonic bioaccumulation of radioactivity, and ion-exchange mechanisms involving the particulate fallout, the soluble elements in the river water, and the membrane surfaces of the organisms. These processes would all occur simultaneously, but the sudden appearance of fresh fallout in *Rangia* makes it seem probable that the fallout particles are filtered directly as they settle to the bottom of the river. Certain more soluble nuclides, such as ^{137}Cs and $^{103-106}$Ru, might preferentially dissolve as the particles settle.

The concentration of total fallout activity increased downstream from Wilson Creek to Union Point on May 18, and, 2 weeks later, clams from Pinecliff contained less total activity than did clams farther downstream at Cedar Point (Table 2). This relation might suggest that fallout radioactivity enters the Trent-Neuse ecosystem not as uniform fallout on the surface of the water but as a discrete "slug" of activity which moves along the stream bed. On May 18, the radioactivity might have been centered around Union Point after having already passed Wilson Creek and Brice Creek, and 2 weeks later, have moved downstream past Pinecliff to the vicinity of Cedar Point. A less simple but more likely explanation for the observed distribution of radioactivity in molluscs along the river is that fallout is more or less uniform, and is in addition slowly carried downstream in the river in a soluble or very finely divided particulate state. As the fresh water of the Trent mixes with the more saline waters of the Neuse estuary, the radioactivity may flocculate or become associated with larger suspended particles which were most suitable for filtering by *Rangia* under the particular biogeochemical conditions at Union Point on and before May 18. Accumulation at downstream stations would be affected also by local currents which could direct more or less filterable particulate matter into any particular sampling area.

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